

Attorney Docket No. SABI-30144 (STC-03-0004)
Application No. 10/632,254
Amendment and Response

In the Claims

Please amend claims 1, 3, 14 and 22, and add new claims 23-24, as shown below.
Please cancel claims 6 and 10.

1. (Currently Amended) A method of preparing a xylene product comprising:

providing a reactor containing a phosphorus-treated ZSM-5-type zeolite catalyst;

initiating start-up of a toluene methylation reaction by contacting the catalyst with a toluene/methanol feed and a cofeed of hydrogen introduced into the reactor at start-up conditions wherein the toluene/methanol feed is introduced into the reactor at a liquid hourly space velocity (LHSV) of ~~from about 2 hr⁻¹ or more~~ than 5 hr⁻¹ and the cofeed of hydrogen is introduced at a hydrogen/(toluene + methanol) ~~hydrogen/hydrocarbon (H₂/HC)~~ molar ratio of less than about 8, ~~and wherein the temperature is from about 500 °C to about 700 °C;~~

operating the reactor at the start-up conditions for about one-half to about 20 hours; and then

operating the reactor at run conditions wherein the LHSV is reduced by at least 5 hr⁻¹ or more from the start-up LHSV to is from to a run LHSV of 10 hr⁻¹ or less and the hydrogen/(toluene + methanol) H₂/HC molar ratio is at least 1.0 ~~and the temperature is from about 500 °C to about 700 °C.~~

2. (Original) The method of claim 1, wherein:

the phosphorus-treated ZSM-5-type zeolite catalyst having a total phosphorus content of from about 0.01 g P/g zeolite to about 0.15 g P/g zeolite.

Attorney Docket No. SABI-30144 (STC-03-0004)
Application No. 10/632,254
Amendment and Response

3. (Currently Amended) The method of claim 1, wherein:

the start-up LHSV is from about 10 hr⁻¹ to about 50 hr⁻¹ ~~or less~~.

4. (Original) The method of claim 1, wherein:

the para-xylene content is at least 90% in the xylene product.

5. (Currently Amended) The method of claim 1, wherein:

the start-up H₂/HC hydrogen/(toluene + methanol) molar ratio is from about 0.1 to about 8.0.

6. (Original) The method of claim 1, wherein:

the reactor is operated at a pressure of from about 10 to about 50 psig.

7. (Original) The method of claim 1, wherein:

the toluene/methanol feed has a toluene/methanol molar ratio of from about 1:2 to about 10:1.

8. (Original) The method of claim 1, wherein:

the ZSM-5-type zeolite catalyst is treated with at least one of phosphoric acid and ammonium hydrogen phosphate.

9. (Original) The method of claim 1, wherein:

Attorney Docket No. SABI-30144 (STC-03-0004)
Application No. 10/632,254
Amendment and Response

the reactor temperature is initially from 200 °C or above and upon introduction of the toluene/methanol feed the reactor temperature is gradually increased at a rate of 1 to 10 °C/min to final start-up temperature from about 500 °C to about 700 °C.

10. (Canceled)

11. (Original) The method of claim 1, wherein:

the catalyst exhibits stable activity for at least 25 hours after start-up of the toluene methylation reaction.

12. (Original) The method of claim 1, wherein:

the catalyst has a silica/alumina mole ratio prior to phosphorus treatment from about 25 to about 300.

13. (Original) The method of claim 1, wherein:

there is substantially no structural aluminum loss of the catalyst during the toluene methylation reaction.

Attorney Docket No. SABI-30144 (STC-03-0004)
Application No. 10/632,254
Amendment and Response

14. (Currently Amended) A method of preparing a xylene product comprising:

providing a reactor containing a phosphorus-treated ZSM-5-type zeolite catalyst using a silica/alumina mole ratio of from 25 to 300 prior to phosphorus treatment and a total phosphorus content of from about 0.01 g/g zeolite to about 0.15 g/g zeolite;

initiating start-up of a toluene methylation reaction by contacting the catalyst with a toluene/methanol feed and a cofeed of hydrogen introduced into the reactor at start-up conditions wherein the toluene/methanol feed is introduced into the reactor at a liquid hourly space velocity (LHSV) of from about $\pm 10 \text{ hr}^{-1}$ to about ~~50~~ 90 hr^{-1} and the cofeed of hydrogen is introduced at a hydrogen/(toluene + methanol) ~~hydrogen/hydrocarbon~~ (H_2/HC) molar ratio of less than about ~~8~~ 5 and ~~wherein the temperature is from about 500 °C to about 700 °C;~~

operating the reactor at the start-up conditions for about one to about five hours; and then

operating the reactor at run conditions wherein the LHSV is reduced by 5 hr^{-1} or more from the start-up LHSV to a run LHSV of 10 from 50 hr^{-1} or less and the hydrogen/(toluene + methanol) H_2/HC molar ratio is increased from that of the start-up conditions at least 5 and the temperature is from about 500 °C to about 700 °C.

15. (Currently Amended) The method of claim 14, wherein:

the start-up hydrogen/(toluene + methanol) H_2/HC molar ratio is from about 0.1 to about 8.0.

Attorney Docket No. SABI-30144 (STC-03-0004)
Application No. 10/632,254
Amendment and Response

16. (Original) The method of claim 14, wherein:

the reactor is operated at a pressure of from about 10 to about 50 psig.

17. (Original) The method of claim 14, wherein:

the toluene/methanol feed has a toluene/methanol molar ratio of from about 1:2 to about 10:1.

18. (Original) The method of claim 14, wherein:

the ZSM-5-type zeolite catalyst is treated with phosphoric acid or ammonium hydrogen phosphate.

19. (Original) The method of claim 14, wherein:

the reactor temperature is initially from 200 °C or above and upon introduction of the toluene/methanol feed the reactor temperature is gradually increased at a rate of 1 to 10 °C/min to final start-up temperature from about 500 °C to about 700 °C, and maintaining the reactor temperature from about 500 °C to about 700 °C.

20. (Original) The method of claim 14, wherein:

the catalyst exhibits stable activity for at least 500 hours after start-up of the toluene methylation reaction.

Attorney Docket No. SABI-30144 (STC-03-0004)
Application No. 10/632,254
Amendment and Response

21. (Original) The method of claim 14, wherein:

the para-xylene content is at least 90% in xylene product.

22. (Currently Amended) A method of preparing a xylene product comprising:

providing a reactor containing a non-steamed, phosphorus-treated ZSM-5-type zeolite catalyst using a silica/alumina mole ratio of from 25 to 300 prior to phosphorus treatment and a total phosphorus content of from about 0.02 g/g zeolite to about 0.13 g/g zeolite;

initiating start-up of a toluene methylation reaction by contacting the catalyst with a toluene/methanol feed and a cofeed of hydrogen introduced into the reactor at start-up conditions wherein the toluene/methanol feed is introduced into the reactor at a liquid hourly space velocity (LHSV) of from about $\pm 10 \text{ hr}^{-1}$ to about 50 hr^{-1} and the cofeed of hydrogen is introduced at a hydrogen/(toluene + methanol) hydrogen/hydrocarbon (H_2/HC) molar ratio of less than about 8, and wherein the temperature is from about 500 °C to about 700 °C;

operating the reactor at the start-up conditions for about one to about two hours; and then

operating the reactor at run conditions wherein the LHSV is ~~from 5 hr^{-1} or less~~ reduced by 10 hr^{-1} or more from the start-up LHSV to a run LHSV of 10 hr^{-1} or less and the hydrogen/(toluene + methanol) H_2/HC molar ratio is increased by at least 2 and the temperature is from about 500 °C to about 700 °C; and wherein

the catalyst exhibits stable activity for at least 500 hours after start-up of the toluene methylation reaction.

Attorney Docket No. SABI-30144 (STC-03-0004)
Application No. 10/632,254
Amendment and Response

23. (New) The method of claim 1, wherein:

the hydrogen/(toluene + methanol) molar ratio is increased by at least about 2
when switching from start-up conditions to run conditions.

24. (New) The method of claim 14, wherein:

the hydrogen/(toluene + methanol) molar ratio is increased by at least about 2
when switching from start-up conditions to run conditions.